

- 11 -

## WHAT IS CLAIMED IS:

1. A method of making multiple carbonaceous nanomaterials, comprising:  
introducing at least one carbon source, at least one nitrogen source,  
5 at least one copper source, at least one yttrium source, at least one transition metal, and at least one trimetasphere-forming metal into a reaction chamber; and  
reacting the at least one carbon source, nitrogen source, copper source, yttrium source, transition metal, and trimetasphere-forming metal  
10 under conditions effective to produce a reaction product comprising trimetaspheres, nanotubes, and at least one of (i) fullerenes and (ii) metallofullerenes.
2. The method of claim 1, wherein the reaction product comprises  
15 trimetaspheres, nanotubes, fullerenes, and metallofullerenes.
3. The method of claim 1, wherein the carbon source is graphite.
4. The method of claim 3, wherein the graphite is graphite  
20 powder.
5. The method of claim 3, wherein the at least one carbon source is a container composed of graphite, and the at least one nitrogen source, copper source, yttrium source, transition metal, trimetasphere-forming metal  
25 and optionally carbon are placed inside the container which is placed in the reaction chamber.
6. The method of claim 5, wherein the container is a cored  
graphite rod.

30

- 12 -

7. The method of claim 1, wherein the at least one nitrogen source is N<sub>2</sub> gas.

8. The method of claim 7, wherein the N<sub>2</sub> gas and helium are  
5 supplied into the reaction chamber.

9. The method of claim 8, wherein the helium is supplied into the reaction chamber at a flow rate of about 800 ml/min to about 1200 ml/min, the N<sub>2</sub> is supplied into the reaction chamber at a flow rate of about 80 ml/min  
10 to about 120 ml/min, and gas pressure within the reaction chamber is about 800 torr to about 1000 torr.

10. The method of claim 1, wherein the at least one nitrogen source is at least one nitride material.  
15

11. The method of claim 1, wherein the at least one copper source is Cu and/or at least one copper-containing compound.

12. The method of claim 1, wherein the at least one transition  
20 metal is effective to catalyze the formation of the nanotubes.

13. The method of claim 12, wherein the transition metal is Ni and/or Fe.

25 14. The method of claim 1, wherein the at least one yttrium source is Y and/or at least one yttrium-containing compound.

15. The method of claim 1, wherein the trimetasphere-forming metal is selected from the elements of Group IIIB of the periodic table and  
30 the rare-earth metals.

- 13 -

16. The method of claim 1, wherein the at least one trimetasphere-forming metal is yttrium.

5 17. The method of Claim 1, further comprising, prior to the introducing, heating the at least one carbon source, nitrogen source, copper source, yttrium source, transition metal and trimetasphere-forming metal in a vessel at a temperature of about 700°C to about 1100°C and for a period of about 1 hour to about 24 hours.

10 18. The method of claim 1, wherein the at least one transition metal is Ni, the at least one trimetasphere-forming metal is Y, the at least one copper source is Cu, and the at least one nitrogen source is N<sub>2</sub>.

15 19. The method of claim 18, wherein the Ni, Y and Cu have a respective weight ratio of about 0.7/0.55/0.55.

20 20. The method of claim 18, wherein the Ni, Y and Cu have a respective weight ratio of about 0.7/1.65/0.55.

21. The method of claim 18, wherein the carbon source comprises carbon, and the carbon, Ni, Y and Cu have a respective weight ratio of about 1.0/0.7/0.55/0.55.

22. The method of claim 18, wherein the carbon source comprises carbon, and the carbon, Ni, Y and Cu have a respective weight ratio of about 1.0/0.7/1.65/0.55.

23. The method of claim 1, wherein the reaction chamber is in an electric-arc discharge generator.

30

24. The method of claim 1, further comprising, prior to the supplying, determining relative masses of at least the at least one copper

- 14 -

source, yttrium source, transition metal, and trimetasphere-forming metal effective to produce a selected reaction product.

25. A reaction product produced by the method according to claim  
5 1.

26. The reaction product of claim 25, which comprises trimetaspheres, nanotubes, fullerenes, and metallofullerenes.

10 27. A method of making multiple carbonaceous nanomaterials, comprising:  
introducing into a reaction chamber at least one copper source, at least one yttrium source, at least one transition metal, and at least one trimetasphere-forming metal in a carbonaceous material; and  
15 vaporizing the at least one copper source, yttrium source, transition metal, trimetasphere-forming metal and carbonaceous material in the presence of nitrogen and producing a reaction product comprising trimetaspheres, nanotubes, and at least one of (i) fullerenes and (ii) metallofullerenes.

20 28. The method of claim 27, wherein the reaction product comprises trimetaspheres, nanotubes, fullerenes, and metallofullerenes.

25 29. The method of claim 27, wherein the carbonaceous material is graphite.

30 30. The method of claim 27, further comprising placing a carbon source in the carbonaceous material.

30 31. The method of claim 27, wherein the carbonaceous material is a cored graphite rod.

- 15 -

32. The method of claim 27, wherein the nitrogen is N<sub>2</sub> gas.

33. The method of claim 32, wherein the N<sub>2</sub> gas and helium are supplied into the reaction chamber.

5

34. The method of claim 33, wherein the helium is supplied into the reaction chamber at a flow rate of about 800 ml/min to about 1200 ml/min, the N<sub>2</sub> is supplied into the reaction chamber at a flow rate of about 80 ml/min to about 120 ml/min, and gas pressure within the reaction chamber is about  
10 800 torr to about 1000 torr.

35. The method of claim 27, wherein the nitrogen is at least one nitride material which is placed in the carbonaceous material.

15

36. The method of claim 27, wherein the at least one copper source is Cu and/or at least one copper-containing compound.

37. The method of claim 27, wherein the at least one transition metal is effective to catalyze the formation of the nanotubes.

20

38. The method of claim 37, wherein the at least one transition metal is Ni and/or Fe.

39. The method of claim 27, wherein the at least one yttrium  
25 source is Y and/or at least one yttrium-containing compound.

40. The method of claim 27, wherein the at least one trimetaspere-forming metal is selected from the elements of Group IIIB of the periodic table and the rare-earth metals.

30

41. The method of claim 40, wherein the at least one trimetaspere-forming metal is Y.

- 16 -

42. The method of Claim 27, further comprising, prior to the introducing, heating the at least one copper source, yttrium source, transition metal and trimetasphere-forming metal in a vessel at a temperature of about  
5 700°C to about 1100°C and for a period of about 1 hour to about 24 hours.

43. The method of claim 27, wherein the at least one transition metal is Ni, the at least one trimetasphere-forming metal is Y, the at least one copper source is Cu, and the nitrogen is N<sub>2</sub>.  
10

44. The method of claim 43, wherein the Ni, Y and Cu have a respective weight ratio of about 0.7/0.55/0.55.

45. The method of claim 43, wherein the Ni, Y and Cu have a  
15 respective weight ratio of about 0.7/1.65/0.55.

46. The method of claim 43, further comprising introducing carbon in the carbonaceous material, the carbon, Ni, Y and Cu having a respective weight ratio of about 1.0/0.7/0.55/0.55.  
20

47. The method of claim 43, further comprising introducing carbon in the carbonaceous material, the carbon, Ni, Y and Cu having a respective weight ratio of about 1.0/0.7/1.65/0.55.

48. The method of claim 27, wherein the reaction chamber is in an electric-arc discharge generator and the reacting comprises vaporizing the carbonaceous material and the at least one copper source, yttrium source, transition metal, and trimetasphere-forming metal.  
25

49. A reaction product produced by the method according to claim  
30 27.

- 17 -

50. The reaction product of claim 49, which comprises trimetaspheres, nanotubes, fullerenes and metallofullerenes.

51. A method of making multiple carbonaceous nanomaterials,  
5 comprising:  
introducing a carbonaceous material and a powder mixture of at least one copper source, at least one yttrium source, at least one transition metal, and at least one trimetasphere-forming metal into a reaction chamber; and  
reacting the carbonaceous material and powder mixture in the  
10 presence of a gas mixture of nitrogen and helium to produce a reaction product comprising trimetaspheres, nanotubes, and at least one of (i) fullerenes and (ii) metallofullerenes.

52. The method of claim 51, wherein the reacting comprises  
15 vaporizing the carbonaceous material and powder mixture, the vapor condensing.

53. The method of claim 51, wherein the reaction product  
comprises trimetaspheres, nanotubes, fullerenes and metallofullerenes.  
20

54. A method of making trimetaspheres, comprising:  
introducing at least one metal, carbon, nitrogen and copper into a reaction chamber; and  
reacting the at least one metal, a carbon, nitrogen and copper to  
25 produce trimetaspheres.

55. The method of claim 54, wherein the at least one metal is selected from the group consisting of elements of group IIIB of the periodic table and rare earth metals.  
30

56. The method of claim 54, wherein the nitrogen is a nitrogen-containing gas.

- 18 -

57. A reaction product comprising trimetaspheres, nanotubes, and at least one of (i) fullerenes and (ii) metallofullerenes.

5 58. The reaction product of claim 57, comprising trimetaspheres, nanotubes, fullerenes and metallofullerenes.

59. The reaction product of claim 57, which is soot formed by an arc discharge technique.

10

60. A method of making carbon nanotubes, comprising reacting at least a transition metal, yttrium source and carbon in the presence of a nitrogen-containing gas and helium.